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Aircraft pollution – a futuristic view

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Abstract. Impacts of NO_x , H_2O and aerosol emissions from a projected 2050 aircraft fleet are investigated using the Oslo CTM2, with emissions provided through the EU project SCENIC. The aircraft emission scenarios consist of emissions from subsonic and supersonic aircraft. In particular it is shown that aerosol emissions from such an aircraft fleet can have a relatively large impact on ozone, and possibly reduce the total atmospheric NO_x by more than what is emitted by aircraft. Without aerosol emissions this aircraft fleet leads to similar NO_x increases for subsonic (at 11–12 km) and supersonic (at 18–20 km) emissions, 1.35 ppbv and 0.83 ppbv as annual zonal means, respectively. H_2O increases are also comparable at these altitudes: 630 and 599 ppbv, respectively. Tropospheric ozone increases are about 10 ppbv in the Northern Hemisphere due to emissions from subsonic aircraft. Increased ozone loss from supersonic aircraft at higher altitudes leads to ozone reductions of about 39 ppbv in the Northern Hemisphere and 22 ppbv in the Southern Hemisphere. The latter reduction is a result of transport of ozone depleted air from northern latitudes. When including aircraft aerosol emissions, NO_x is reduced due to heterogeneous chemistry. The reduced NO_x seems to counterweight the reduction of ozone from emissions of NO_x and H_2O above 20 km. At these altitudes the NO_x (and thus ozone loss) reduction is large enough to give an aircraft emissions induced increase in ozone. In the height range 11–20 km altitude, however, ozone production is reduced. Heterogeneous reactions and reduced NO_x enhances ClO, further enhancing ozone loss in the lower stratosphere. This results in a 14 ppbv additional reduction of ozone. Although supersonic aircraft have opposite effects on ozone in the upper and lower stratosphere, the change in ozone columns is clearly dominated by the upper stratospheric loss, thus supersonic aircraft aerosol emissions lead to enhanced ozone columns. The largest in-

crease in the ozone column due to aerosol emissions is therefore seen in the Northern Hemispheric autumn and winter, giving a column increase of 4.5 DU. It is further found that at high northern latitudes during spring the heterogeneous chemistry on PSCs is particularly efficient, thereby increasing the ozone loss.

1 Introduction

Air traffic is continually increasing. According to the Global Market Forecast 2004–2023 (Airbus, 2004), revenue passenger kilometres will increase by 5.3% annually in this period, while freight tonne kilometres (air-freight) will increase by 5.9% annually. Although less fuel-efficient aircraft eventually will be replaced with new aircraft, the amount of burnt fuel will increase. A further increase in air traffic is assumed to take place after 2020, and by 2050 new high speed civil transport aircraft (so-called supersonic aircraft) are suggested to be developed to meet the demands for faster transport. While subsonic aircraft will have cruising altitudes up to about 12 km, supersonic aircraft will fly up to altitudes of almost 20 km. In the EU project SCENIC (Scenario of aircraft Emissions and Impact studies on Chemistry and climate, Rogers (2005)), several possible aircraft scenarios for 2050 were developed by Airbus (Rogers, 2005, and C. Marizy, personal communication), covering both commercial and non-commercial traffic.

Cruising at 16–18 km altitude, supersonic aircraft emissions mostly take place well above the tropopause, a region of more efficient accumulation of pollution. This is significantly different from subsonic traffic, with emissions often occurring in the upper troposphere, except for high latitudes where the lower stratospheric emissions are frequent.

Emissions from aircraft include ozone precursors (e.g. NO_x) and depleting substances (e.g. H_2O) as well as aerosol particles, which all affect atmospheric chemistry, e.g. ozone

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production and destruction. Also components important for climate, CO₂ in addition to H₂O are emitted from aircraft. Several studies focusing on aircraft NO_x emissions have been carried out (Johnston, 1971; Hidalgo and Crutzen, 1977; Johnson et al., 1992; Schumann, 1997; Dameris et al., 1998; Grewe et al., 2002; Gauss et al., 2006a), most of them addressing the impact of subsonic aircraft. The atmospheric impact of aircraft emissions has also been thoroughly addressed by e.g. Stolarski et al. (1995), Kawa et al. (1998) Fabian and Kärcher (1997), Brasseur et al. (1998), Isaksen (2003). This study is an extension of previous studies done within the EU projects TRADEOFF, CRYOPLANE and SCENIC. We focus on the atmospheric impact of a future fleet comprising sub- and supersonic aircraft (referred to as a “mixed fleet”). The adopted emissions of the mixed fleet include aerosol particles. In Sect. 2 the model setup and the emission scenarios are described, and in Sect. 3 the atmospheric impact of NO_x and H₂O aircraft emissions is presented. The impact of aerosol emissions from a mixed fleet is studied in Sect. 4, and discussion and conclusions are given in Sect. 5.

2 Model description

The Oslo CTM2 is a global chemical transport model. Advective transport is treated by the highly accurate and low-diffusive Second Order Moments scheme (Prather, 1986), while the Tiedke mass flux scheme (Tiedke, 1989) is used for deep convection. Boundary layer mixing is treated according to the Holtslag K-profile scheme (Holtslag et al., 1990). The model includes comprehensive schemes for tropospheric and stratospheric chemistry, and comprises 98 species whereof 77 are transported. It is designed to study the upper troposphere and the lower stratosphere (UTLS), which is the region that cover the major cruising altitudes of aircraft. The numerical integration of chemical compounds is done applying the Quasi Steady State Approximation (QSSA) (Hestvedt et al., 1978), with three different integration methods depending on the chemical lifetime of the species.

The meteorological data driving the Oslo CTM2 is taken from the European Centre for Medium-Range Weather Forecasts (ECMWF) operational Integrated Forecast System (using 4D-var assimilation) for the year 2000, and are forecasts produced with 12 h of spin-up starting from an analysis at noon on the previous day. The meteorological data are updated every third hour to ensure more realistic transport. The spectral resolution applied is T21 (5.6×5.6 degrees) in 40 vertical layers, extending from the surface to 2 hPa (pressure center of the top model layer at 10 hPa) in σ -pressure hybrid coordinates. The choice of horizontal resolution was made due to the large number of scenarios and the long spin-up time required for stratospheric studies, combined with the computational cost. Vertical model resolution in the UTLS varies between 0.8 and 1.2 km, depending on latitude and

season. At altitudes above 100 hPa the vertical resolution is 20 hPa, corresponding to ~2 km at 18 km altitude.

2.1 Model performance

With a focus on the tropospheric chemistry, the Oslo CTM2 has previously been tested and applied in various works (Berntsen and Isaksen, 1997; Grini et al., 2002; Brunner et al., 2003, 2005; Isaksen, 2003; Isaksen et al., 2005). The stratospheric chemistry module was originally developed by Stordal et al. (1985), and improved by adding heterogeneous chemistry to the scheme (Isaksen et al., 1990). The stratospheric chemistry was introduced in a stratospheric 3-D CTM by Rummukainen et al. (1999), and later in the Oslo CTM2, which already included comprehensive tropospheric chemistry (Gauss, 2003). The model version with both tropospheric and stratospheric chemistry has previously been validated against measurements by satellites, sondes and lidar (Braathen et al., 2003), against satellite (Eskes, 2003; Brunner et al., 2005) and aircraft measurements (Isaksen, 2003). It is found to reproduce ozone measurements well. Further, the Oslo CTM2 has been used to study past and future changes in tropospheric and lower stratospheric ozone (Gauss et al., 2003c, 2006b) as input to studies of radiative forcing, and to study the impact of aircraft emissions of water vapor (Gauss et al., 2003b) and nitrogen oxides (Gauss et al., 2006a).

Recently the Oslo CTM2 (including stratospheric and tropospheric chemistry) has been improved by a new heterogeneous and microphysics scheme which is described by Søvde et al. (2007)¹. It is found to reproduce ozone depletion events like the Ozone Hole and miniholes. An extensive evaluation of the Oslo CTM2 is also given by Søvde et al. (2007)¹, evaluating the model against aircraft, sonde and satellite measurements. Aircraft (Mozaic) flight measurements of ozone are found to be well reproduced on hourly basis for both T42 and T21 horizontal resolution, although T42 performed best. Furthermore, sonde and satellite measurements of ozone were reproduced well. The conditions where T21 performed significantly poorer than T42 was in Ozone Hole conditions where the averaging of polar grids cause too much diffusion across the polar vortex. Comparisons of the Oslo CTM2 with a number of vertical profiles from SAGE II for a standard 2000 run are shown for two latitude intervals (30 N–45 N and 60 N–75 N, respectively) in Figs. 1a and b. The modelled profiles are taken at the same hour and location (grid box value) as the measurements, showing that ozone is well reproduced in the UTLS. Due to the intermediate lifetime of ozone in the UTLS region, accurate transport as well as chemistry is needed to reproduce measurements. The evaluation of the Oslo CTM2 presented here shows that the Oslo CTM2 performs well.

¹ Søvde, O. A., Gauss, M., Smyshlyaev, S., and Isaksen, I. S. A.: The cold Arctic winter stratosphere of 2004/2005 – a Global Chemical Transport Model study, *J. Geophys. Res.*, to be submitted, 2007.

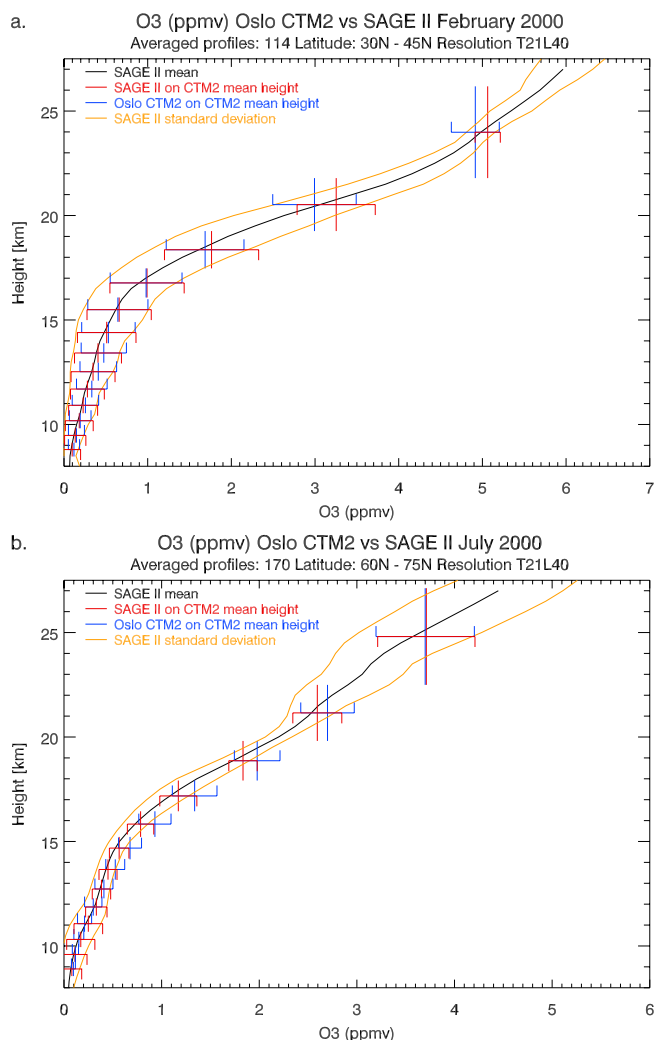


Fig. 1. Evaluation of Oslo CTM2 against SAGE II between 8 and 28 km altitude. Monthly mean ozone for (a) February 2000 between 30°N and 45°N, and (b) July 2000 between 60°N and 75°N. The average SAGE II ozone profile is given as the black line, and its standard deviations as the orange lines. The blue vertical lines are the Oslo CTM2 averages on each model level, and red is SAGE II averaged on the same model levels. Standard deviations are given as horizontal bars for each model level.

A parameter closely connected to transport and the Brewer–Dobson circulation is the age of stratospheric air (Hall and Plumb, 1994). Monge-Sanz et al. (2007) used the ECMWF operational (4D-var) data extending to 0.1 hPa. The age of air for was improved when forecasts on 3 h time resolution was used. For the 40-layer version of meteorological data applied here, we calculated the mean age of air from the age spectrum (according to Hall et al. (1999)), using the applied 2000 meteorology perpetually for 20 years, giving a zonal mean age maximum of 5.7 years at the top of the model domain (Fig. 2). The values at 20 km vary from 4.2 years at

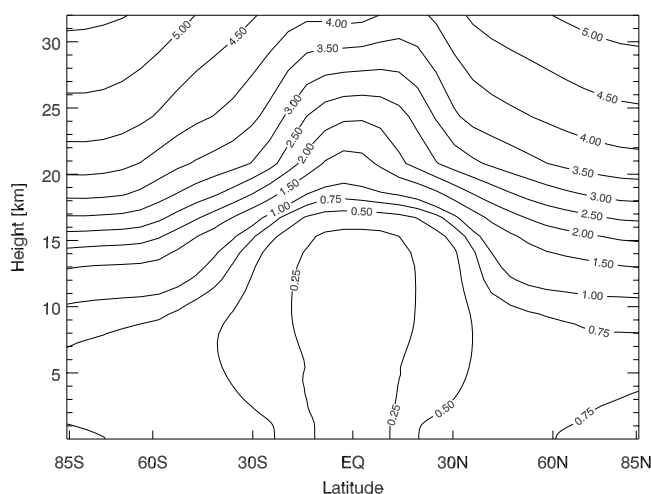


Fig. 2. Mean age of air for the Oslo CTM2 in T21 horizontal resolution, calculated using year 2000 meteorology. See text for details.

the South Pole to 1.3 in the tropics and 3.8 years at the North Pole, corresponding closely to the observed values shown by Monge-Sanz et al. (2007). Ideally the meteorological data should cover the whole stratosphere to assess the age of air. However, based on the calculated age presented here combined with the ability of reproducing measurements, we are confident that transport processes are well represented.

Our confidence is confirmed by looking at the long-lived species, and in Fig. 3 the modelled annual mean N_2O is compared to the UARS Reference Atmosphere Project climatology available from their web-site <http://code916.gsfc.nasa.gov/>. The main structure of the atmospheric circulation with upwelling in the tropics and subsidence at high latitudes is covered, as well as the N_2O levels. Modelled gradients at low to mid latitudes above 20 km are somewhat underestimated, which is partly due to the coarse model vertical resolution at the highest altitudes, but also due to the upper boundary values, which are too low at low latitudes and too high at high latitudes. In this way reproduction of steep gradients observed is hindered by the boundary conditions, which prevents downward transport of low N_2O at high latitudes. All in all, the model is reproducing measurements well.

2.1.1 Emission set-up

The natural and industrial emissions are taken from the EU project POET (Precursors of Ozone and their Effects in the Troposphere) (POET report, 1999). The model is set up to simulate the year 2050 according to the SCENIC project (Rogers, 2005), where the industrial emissions of NO_x , CO and volatile organic compounds (VOC) are scaled up from the year 2000 by 2.2, 1.53 and 1.55, respectively. As the initial chemical conditions, we use a steady-state atmospheric composition from a calculation of a previous 2050 study,

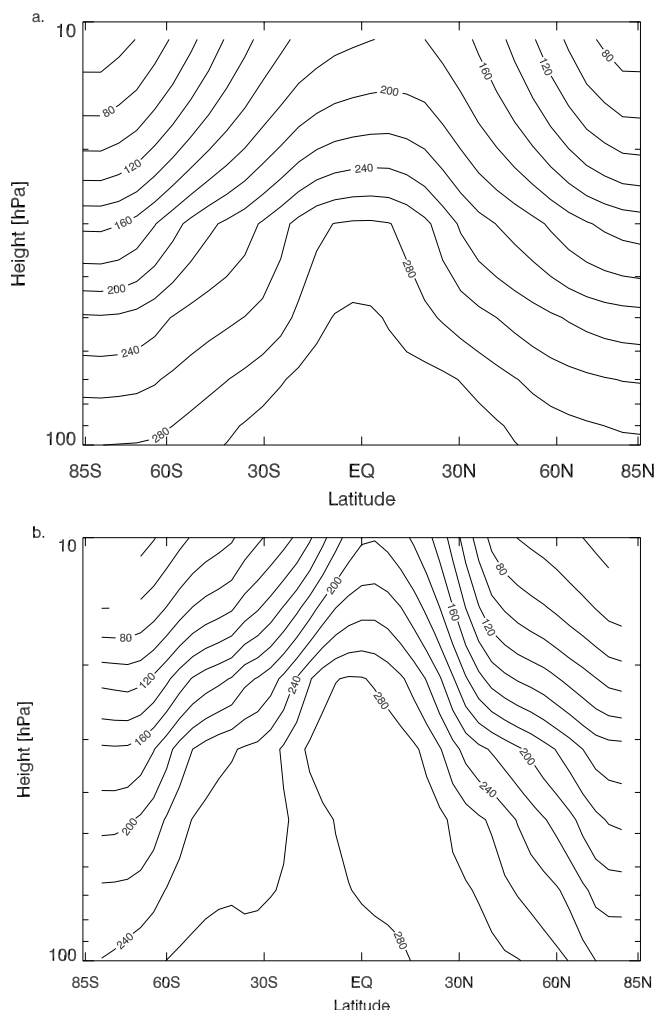


Fig. 3. Annual mean N_2O from (a) the Oslo CTM2 and (b) the UARS Reference Atmosphere Project climatology.

with the same boundary conditions. The boundary conditions in both studies were based on World Meteorological Organization (2003) for longlived species (e.g. CFCs), and methane and N_2O were set to 2549 ppbv and 372 ppbv, respectively, at the surface. Also emissions of stratospheric species were set to 2050 level, as were the model upper boundary conditions for all species (taken from the Oslo 2-D model (Stordal et al., 1985)). Otherwise, natural emissions are set to the level of year 2000. In the Oslo CTM2, tropospheric water vapor is retrieved from the meteorological data, while stratospheric water vapor is calculated assuming that the H_2 content in water vapor, methane and hydrogen gas is constant ($\text{H}_{\text{sum}} = \text{H}_2\text{O} + 2\text{CH}_4 + \text{H}_2$), where only CH_4 is transported. H_{sum} is set to 8.598 ppmv excluding aircraft emissions of H_2O . However, the H_2O emitted from aircraft is transported as a separate species, which is superposed on this calculated value of H_2O . If PSC2 is formed, the solid H_2O

is calculated and removed from the gas phase, and we keep track of the sedimented amount of solid H_2O , thereby taking dehydration into account.

Starting from the initial conditions (the previous 2050 study), a spin-up of 7 years without aircraft emissions was performed, followed by additional six-year runs including each of the aircraft emission scenarios, where results from the last year of the simulations are presented here. The aircraft emission inventories studied in this work are listed in Table 1, and are given in 1×1 degrees horizontal resolution and 305 m vertical resolution. These data are interpolated into the Oslo CTM2 grid each model time step. Due to the relatively coarse horizontal resolution, a plume model is applied to the aircraft emissions (Kraabøl et al., 1999) to include the effects of in-plume chemistry, changing the NO_x partitioning of the emissions. In this way the plume processed emissions can be emitted into the model grid.

Based on a predicted increase in non-commercial as well as commercial aircraft traffic, emissions from the most likely mixed fleet is given in MIX, assuming 501 supersonic aircraft and 114.1 million supersonic passengers per year and a supersonic aircraft cruise speed of 2.0 Mach. Aircraft emissions in this study include NO_x and H_2O , omitting other products of fuel combustion such as CO_2 , CO, different hydrocarbons and sulfur oxides; although CO_2 is an important greenhouse gas, the lifetime is too long to distinguish the aircraft effect from other sources, and increases of the other combustion products are low compared to the background concentrations, hence they are expected to have a small atmospheric impact (Fabian and Kärcher, 1997). The aircraft emissions comprise commercial subsonic, commercial supersonic, general aviation and military fleet, and the respective emissions of NO_x and H_2O are listed in Table 2. H_2O emissions are calculated as 1.25 of fuel burned.

NO_x in the troposphere produces ozone in the presence of peroxy radicals and sunlight, while in the stratosphere it causes ozone depletion through the catalytic NO_x cycle. H_2O is an important source of the OH radical ($\text{H}_2\text{O} + \text{O}^1\text{D} \rightarrow 2\text{OH}$), which contributes to ozone loss. In this study we focus on the total atmospheric impact of NO_x and H_2O emissions. Due to the high background concentration of H_2O in the troposphere, the effects of H_2O emissions in the troposphere are negligible. However, in the stratosphere H_2O is very low, and therefore aircraft emissions have a larger effect on the OH formation.

Scenario AERO is similar to MIX, except that aircraft aerosol emissions are included. Aerosols offer surfaces for heterogeneous chemistry, which is important for NO_x chemistry. The surface area density (SAD) of these aerosols was included in the model as a transported tracer with SAD-emissions calculated according to Danilin et al. (1998),

$$\text{SAD} = \frac{3\mu_f \rho_a \xi \text{EI}(S) \mu_{\text{H}_2\text{SO}_4}}{r \rho_s w_{\text{H}_2\text{SO}_4} \mu_s} \quad (1)$$

Table 1. The aircraft scenarios used in this study.

Scenario	Fleet	NO _x emissions	H ₂ O emissions	Aerosol emissions
noAC	No aircraft	no	no	no
MIX	Sub- and supersonic (mixed) fleet	See Table 2		no
AERO	MIX with aerosol emissions	as MIX	as MIX	yes
noH2O	MIX without H ₂ O emissions	as MIX	no	no
noBACK	MIX without background aerosols	as MIX	as MIX	no

Table 2. The aircraft scenario emissions of the mixed fleet taken from Marizy et al. (2007)^{??}.

Emitted species	Mixed fleet emissions 2050				
	Total	Commercial subsonic	Commercial supersonic	General aviation	Military aviation
NO _x (Tg(NO ₂)/yr	7.445	6.987	0.274	0.074	0.110
H ₂ O (Tg(H ₂ O)/yr	901.0	800.6	74.4	10.6	15.4
Fuel burned (Tg)	720.8	640.5	59.5	8.5	12.3
Aerosols	Calculated by Eq. (1), as described in the text.				

where $\rho_s=1.5\text{ g cm}^{-3}$ is the density of sulfur, ρ_a is the density of air, $w_{\text{H}_2\text{SO}_4}=50\%$ is the weight percent of sulfuric acid and μ_S and $\mu_{\text{H}_2\text{SO}_4}$ are the molecular weights of sulfur and sulfuric acid, respectively. The particle radius is assumed to be $r=10\text{ nm}$, and the emission index of sulfur is $\text{EI}(\text{S})=4\times 10^{-4}$. The fuel mixing ratio (μ_f) is calculated from the MIX NO_x emissions applying the $\text{EI}(\text{NO}_x)$ for supersonic aircraft (4.6 g/kg). Even though subsonic aircraft have a higher emission index for NO_x and may result in a too low SAD value for the subsonic flight levels, the application of the chosen emission index is justified by the uncertainty of $\text{EI}(\text{S})$ and the conversion factor ($\xi=0.05$) of sulfur to sulfate, and the fact that the particles will be most important in the stratosphere. To simulate washout of emitted species, a method suggested by Danilin et al. (1998) is used, where emitted gases and aerosols are removed with e-folding time of 5 days below 400 hPa. In the top model layer, an e-folding time of 1 year is applied to simulate transport and mixing upwards. In this study, sedimentation of aircraft aerosols is not considered, but will be discussed in Sect. 5. Aircraft aerosol emissions are superimposed on the Oslo CTM2 background aerosols, which are taken from a monthly zonal mean SAD climatology derived from various satellite measurements, primarily SAGE II according to Thomason et al. (1997), and data for the year 1999 is applied. A sensitivity study of the MIX scenario without background aerosols (noBACK) is also carried out.

3 Impact of emissions from a mixed fleet

Subsonic and supersonic aircraft cruise at different altitudes. The subsonic fleet is larger than the supersonic fleet, giving the largest contribution to aircraft NO_x emissions. This is clearly seen from Fig. 4, showing the annual zonal mean change in reactive nitrogen (NO_y) with maximum increase at 11 km, and a secondary maximum at about 18 km. Clearly, the secondary maximum is due to supersonic aircraft, while the 11 km maximum is due to subsonic aircraft. The seasonal variation of the monthly zonal mean maximum due to subsonic aircraft ranges from 1.1 in September to 1.9 ppbv in May (Fig. 5). During winter and spring, a larger fraction is emitted into the stratosphere than during summer months due to a lower tropopause, increasing the impact of subsonic aircraft. The higher tropopause in summer is accompanied by relatively strong convection, mixing and washout at mid-latitudes, leading to a reduction in the NO_y increases in the mid-latitude troposphere. This causes the maximum impact of aircraft emissions to shift northward (Kraabøl et al., 2002). A similar latitude shift of the maximum value is reported by Gauss et al. (2003a).

Although the contribution of supersonic aircraft to the zonal mean change is much smaller than for subsonic aircraft, the maximum NO_y changes due to supersonic aircraft are locally comparable with subsonic aircraft emissions, especially in the summer months. Supersonic flights are confined mostly to the North Atlantic corridor, and this small zonal extent results in the slightly lower zonal mean maximum shown in Fig. 4.

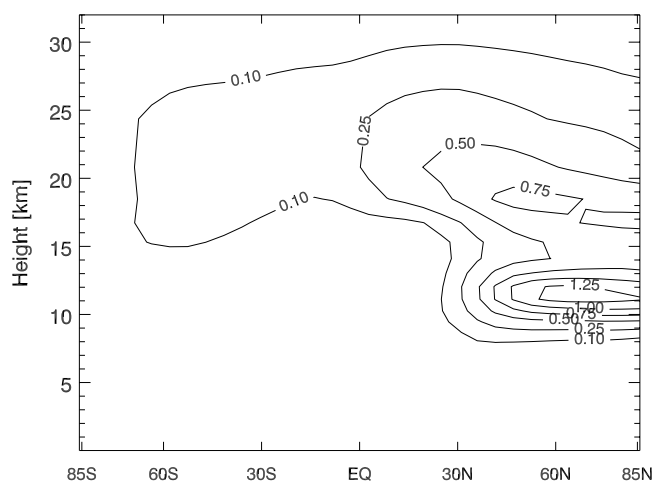


Fig. 4. Effect of mixed fleet aircraft emissions on NO_y , annual zonal mean (MIX minus noAC). Max: 1.35 ppbv.

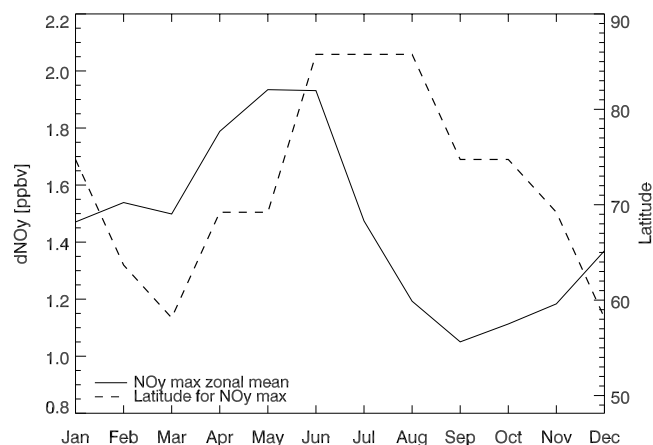


Fig. 5. Latitude and magnitude variation of the monthly mean maximum effect of aircraft emissions on NO_y (MIX minus noAC): 1.1 to 1.9 ppbv.

Aircraft emissions of H_2O contribute to an annual zonal mean increase of up to 624 ppbv above the background H_2O (Fig. 6), ranging from 609 to 833 ppbv in the monthly mean. The maximum change in monthly zonal mean H_2O due to aircraft is mainly located at 11 km altitude, and is largest during summer when the maximum shifts to higher altitudes. As seen before, this shift is caused by higher tropopause levels extending transport and washout processes to higher altitudes.

The annual zonal mean effect of aircraft emissions (NO_x and H_2O) on ozone is given in Fig. 7, showing a maximum of 10 ppbv at 10 km and a minimum of -39 ppbv at 24 km. From month to month, these values vary from 7.2 to 18.4 ppbv (corresponding to a change of 5.5 to 8.9%) and

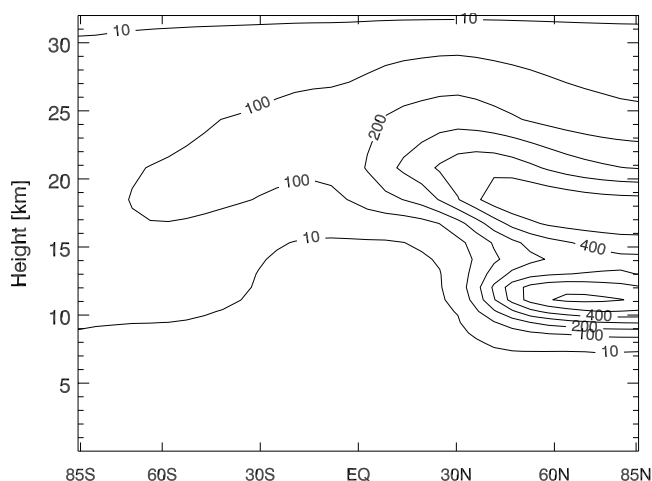


Fig. 6. Effect of aircraft emissions on H_2O , annual zonal mean (MIX minus noAC). Max: 629.69 ppbv.

from -48.8 to -31.4 ppbv (-1.8 to -0.8%), respectively (Fig. 8). The maximum variation largely follows the variation of the NO_y maximum, but at an altitude of 9–10 km. This means that the maximum ozone production is located at a lower altitude than the maximum of emissions. Unlike NO_y and NO_x , the maximum ozone increase is shifted northwards for a longer time period in spring and summer, partly due to a higher tropopause and more washout processes at mid-latitudes. Also important for this shift is the increased amount of sunlight at high latitudes during spring and summer, causing more ozone production even for moderate NO_x increases.

NO_x contributes to ozone production in the presence of sunlight and peroxy radicals. At some altitude (the turn-over point), the amount of these radicals is too low for ozone production, and the catalytic NO_x cycle takes over, reducing ozone. The zero line of Fig. 7 does not represent the turn-over point, since transport processes also affect this figure: Downward transport of ozone depleted air will lower the altitude of the zero line. H_2O emissions, however, contribute to ozone destruction at all altitudes through the OH chemistry. Looking at Fig. 8, the monthly variation of maximum ozone depletion due to aircraft resembles the variation of maximum ozone production. The maximum depletion is located at about 30–40 N, around 24 km, except during summer months, when a shifting northwards is seen, accompanied by a lowering of the maximum depletion to about 20 km altitude. This shift is due to stratospheric transport processes and more sunlight at higher latitudes. Being located above the NO_x and NO_y maxima, the maximum ozone depletion is caused by upward transport of NO_x and H_2O , followed by more effective depletion at higher altitudes. Most of this ozone depletion is due to supersonic aircraft; only a small fraction originates from the upward transport of subsonic aircraft emissions. Mixing

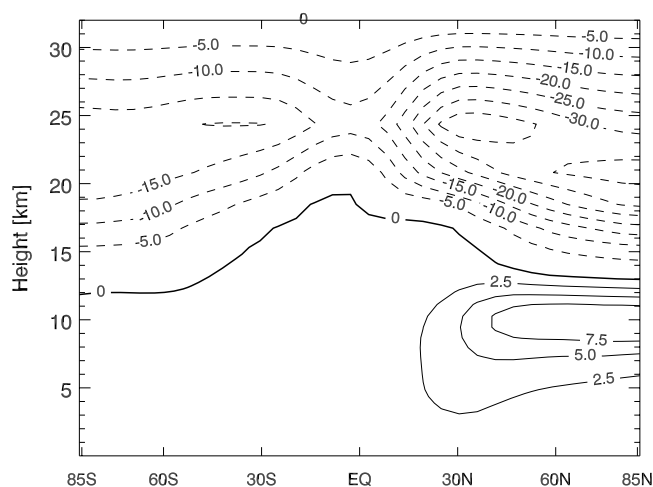


Fig. 7. Effect of aircraft emissions on O_3 , annual zonal mean (MIX minus noAC). Min: -39.24 ppbv, max: 9.96 ppbv.

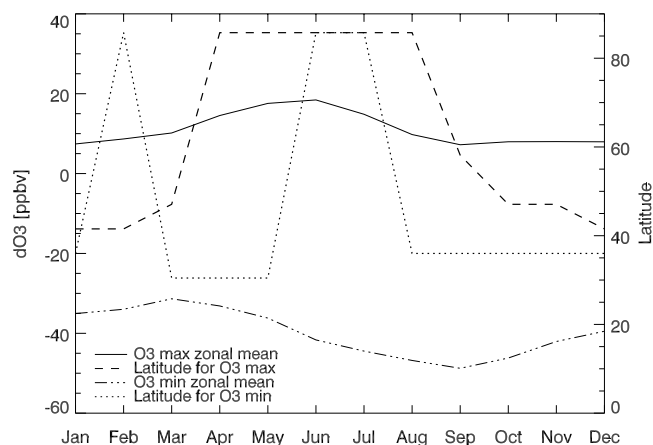


Fig. 8. Magnitude and latitude variation of the monthly zonal mean maximum/minimum effect of aircraft emissions on O_3 (MIX minus noAC), maximum ranging from 7.2 to 18.4 ppbv (due to subsonic aircraft) and minimum from -48.8 to -31.4 ppbv (due to supersonic aircraft).

of tropospheric (ozone increased) and stratospheric (depleted ozone) air in a mixed fleet scenario will therefore lower the tropospheric ozone increase due to subsonic aircraft.

Inter-hemispheric transport of stratospheric aircraft emissions is clearly seen in Figs. 4 and 6, in contrast to transport of subsonic aircraft emissions at lower altitudes, which is very limited (not shown). The high altitude transport will affect ozone in the Southern Hemisphere although emissions occur in the Northern Hemisphere.

When affecting ozone, aircraft emissions may also alter the total ozone column. The changes in daily zonal averages of the total ozone columns for the last two simulated years

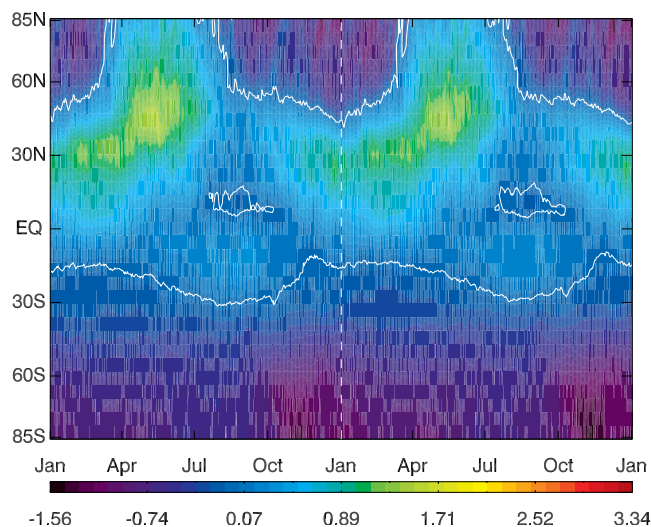


Fig. 9. Changes in the daily zonal mean total ozone column [Dobson Units] due to aircraft emissions (MIX minus noAC) for the last two years of simulation, ranging from -1.56 DU to 1.65 DU.

are shown in Fig. 9, ranging from -1.56 DU to 1.65 DU. The increase in the total column is located in the area of highest flight density. At higher latitudes, however, the supersonic aircraft contribute more to ozone loss, showing a net decrease in the total column, except during spring and summer months where the ozone production maximum is shifted northwards, overriding the ozone loss at higher altitudes. The ozone decrease in the Southern Hemisphere is mainly due to cross-hemispheric transport of emissions from supersonic aircraft, and to some extent transport of ozone depleted air. From August to October, there is also a decrease in total ozone in the tropical region. This is because the tropospheric ozone production due to aircraft emissions is small due to a high tropopause and increased washout of NO_y during these months, causing the stratospheric depletion to dominate.

A sensitivity study omitting aircraft emissions of H_2O (scenario noH2O) reveals that ozone changes due to aircraft emissions are mainly caused by NO_x . This is carried out looking at the MIX – noH2O difference. Emissions of H_2O give maximum ozone losses ranging from 6.7 to 11.4 ppbv in the monthly zonal mean (not shown). The maximum effect of H_2O on ozone is generally located at 25 – 30 N and 24 km height with values ranging from 6 to 7 ppbv. However, the largest decrease in ozone is found in October–November due to PSC chemistry taking place at about 18 km at the South Pole. The ozone reduction due to aircraft emissions of H_2O is most prominent in the stratosphere, although there is some ozone reduction caused by H_2O in the troposphere. At the highest latitudes around 24 km altitude and during summer, H_2O emissions slightly enhance heterogeneous chemistry on background aerosols, reducing NO_x and thereby increasing ozone.

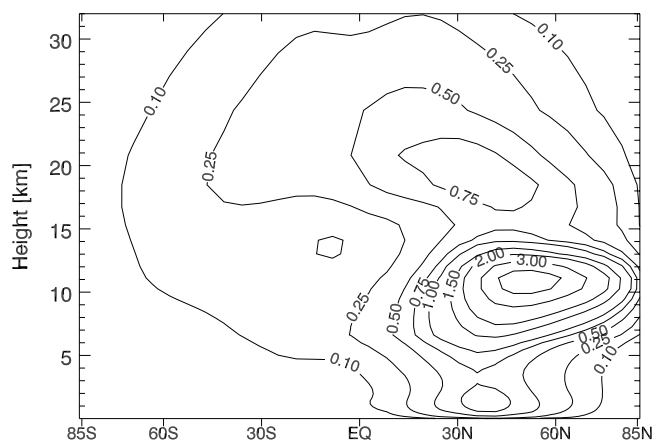


Fig. 10. Annual zonal mean of aircraft aerosol surface area density from the mixed fleet. Max: $4.7 \mu\text{m}^2 \text{cm}^{-3}$.

4 Impact of aerosol emissions from a mixed fleet

Heterogeneous reactions may take place on sulfate aerosols converting NO_x into HNO_3 – reactions being most effective at low temperatures. Depending on the altitude of this conversion, the resulting reductions of NO_x will either reduce the production of ozone in the troposphere and lowermost stratosphere, or reduce the loss at higher altitudes. However, the formation of PSCs and chlorine activation may be enhanced by a reduction of NO_x .

Calculating the aircraft aerosol SAD emissions based on the method of Danilin et al. (1998), we find an annual zonal average SAD maximum of $4.7 \mu\text{m}^2/\text{cm}^3$ (Fig. 10), the monthly zonal mean maximum varying steadily through the year (Fig. 11), ranging from $3.8 \mu\text{m}^2/\text{cm}^3$ in September to $6.2 \mu\text{m}^2/\text{cm}^3$ in May. As for NO_y emissions, the largest values are found in the Northern Hemispheric winter and spring at about 47°N and 11 km height, also shifting slightly to the north during summer and autumn because of the meteorological conditions. Although the maximum value decreases during summer conditions, the surface area density values in the stratosphere increase due to upward transport. The tropospheric reduction of NO_x due to aerosol emissions is largest in spring, while the stratospheric reduction is largest in autumn. Hence, the largest reduction of ozone production due to aerosol emissions from aircraft occurs during spring, while the largest reduction of ozone loss at higher altitudes takes place in autumn.

The heterogeneous conversion of NO_x to HNO_3 will also occur on background aerosols, similarly affecting the production and loss of ozone. In Fig. 12 we see the annual zonal mean effect of aircraft aerosol emissions on ozone (AERO – MIX), superimposed on the effect of background aerosols (MIX – noBACK), for latitudes between 30 and 60 degrees in (a) the Northern and (b) Southern Hemisphere. Most promi-

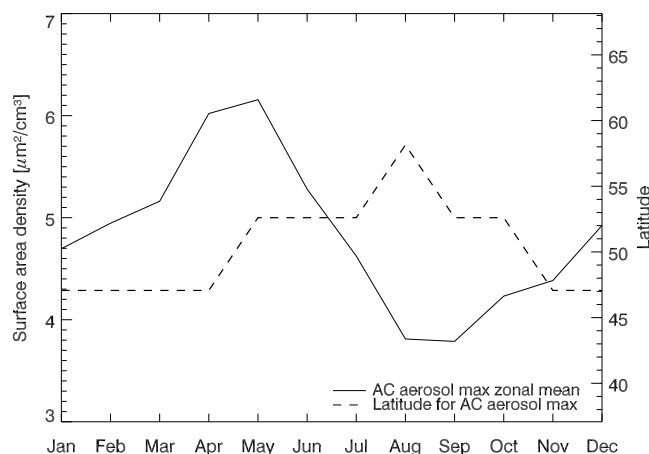


Fig. 11. Variation of the magnitude and latitude of the monthly zonal mean maximum of aircraft aerosol surface area density.

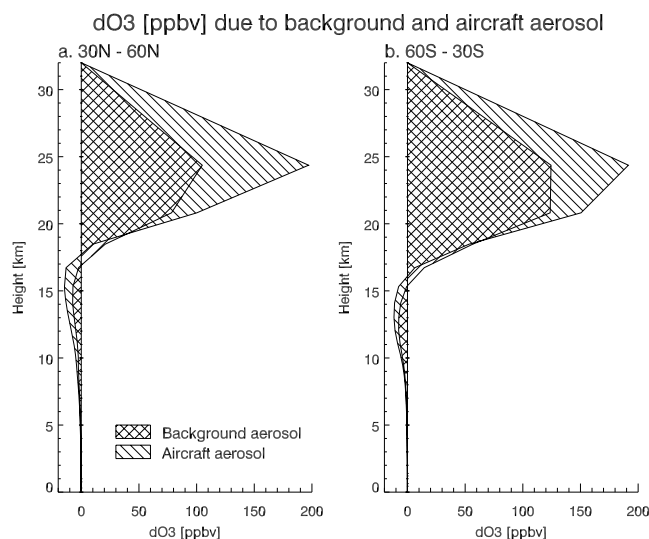


Fig. 12. Annual zonal mean effect of background aerosol on O_3 (cross-hatched) and the contribution from aircraft aerosol emissions (hatched). (a): 30°N – 60°N , (b): 60°S – 30°S (given in ppbv).

nent is the increase in ozone between 18 and 30 km due to ozone loss reduction, where the effect of aircraft aerosols is comparable with the background aerosols. For altitudes between 12 and 20 km aircraft aerosols contribute to an ozone reduction through reduced production and a slight increase in ozone loss due to more PSCs or supercooled ternary solutions (STS).

The amount of PSCs is dependent on HNO_3 and H_2SO_4 , where the latter is calculated from the amount of background aerosols available (described by Søvde et al., 2007¹). Thus an increase in HNO_3 and H_2SO_4 due to aircraft aerosol emissions leads to a slight increase of PSCs and therefore an

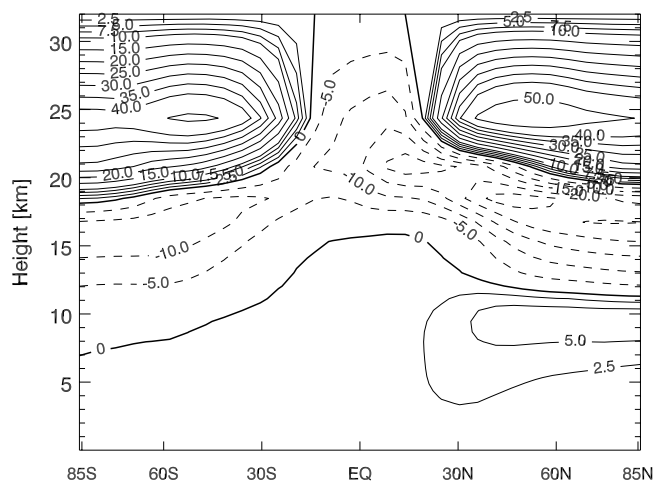


Fig. 13. Annual zonal mean effect of a mixed fleet with aerosol emissions on O_3 (AERO minus noAC), given in ppbv.

increase in ozone loss. Also, a reduction in NO_x will result in more active chlorine available for depleting ozone. These effects are apparent at altitudes between 12 and 20 km.

The maximum increase of ozone for a mixed fleet including aerosol emissions (Fig. 12), has a larger magnitude than the maximum decrease due to NO_x and H_2O emissions only (Fig. 7). Aircraft aerosols give a reduction of NO_x through heterogeneous reactions, reducing the loss of ozone due to the NO_x cycle. The increase in aerosol SAD also intensify the heterogeneous conversions, lowering the concentration of HCl and thus the heterogeneous loss of $ClONO_2$. In this way more NO_x is bound up in $ClONO_2$, while also the ClO is increased, since less NO_2 is available for forming $ClONO_2$. The same applies for BrO/Br ONO_2 . Both NO_x and ClO are responsible for ozone loss, so by looking at the effect on ozone (AERO – noAC, Fig. 13) we see that above 18–20 km the NO_x cycle is more important for ozone than the ClO – giving an increase of ozone due to reduced NO_x . From 18–20 km and down to about 11–12 km (lower at the Southern Hemisphere), the turn-over point where NO_x starts to produce ozone will be reached. However, in this region enhanced ClO and BrO increase the ozone loss in addition to the reduced ozone production due to lower NO_x . Below 11–12 km the NO_x reduction due to aerosols is not large enough to overcome the aircraft emissions of NO_x , resulting in increased ozone production there (Fig. 13). Therefore, a mixed fleet with aerosol emissions results in an increase in the daily zonal mean total column at most latitudes (Fig. 14; AERO minus noAC). At the lowermost latitudes through the year, and at the highest latitudes during summer, the reduced ozone production and enhanced loss due to ClO cause a decrease in ozone. The ozone column reduction at high latitudes during spring is due to increased heterogeneous chemistry on PSCs and therefore increased ozone loss due to ClO.

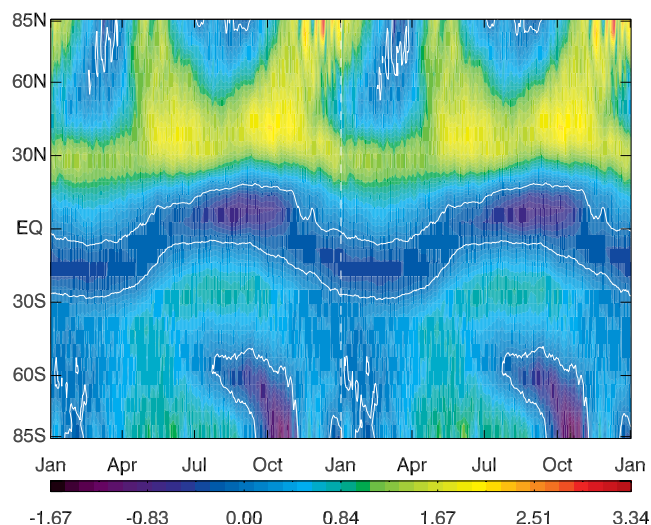


Fig. 14. Changes in the daily zonal mean total ozone column [Dobson Units] due to the mixed fleet aircraft emissions including aerosol emissions (AERO minus noAC), for the last two years of simulations, ranging from -1.67 DU to 3.34 DU.

Separating the effect of aircraft aerosols, by comparing the AERO scenario to the mixed fleet scenario (MIX), aircraft aerosols reduces the total ozone column in spring, and increase it in autumn (Fig. 15). Again, the decrease is due to reduced ozone production below the turn-over point (about 20 km), combined with increased ozone loss by ClO and due to heterogeneous reactions on PSCs/STS. The increase is due to ozone loss decrease at higher altitudes (Fig. 16). The decrease in ozone column near the North Pole during May through June is due to more ozone loss below 20 km (due to ClO increase and reduced ozone production) than the ozone loss is decreased above 20 km (due to NO_x). Aerosols are also transported into the Southern Hemisphere, readily reducing NO_x there as well, giving similar results, however smaller in magnitude. The cross-hemispheric transport is most effective above 18 km, mainly resulting in reduced ozone loss. However, ClO is also increased here, to some extent enhancing the ozone loss below 18 km. The ozone loss is also increased in spring when heterogeneous chemistry on PSCs/STS is enhanced, increasing the Ozone Hole depth.

Stratospheric H_2O increases slightly due to the presence of aircraft aerosols (not shown), with an annual zonal mean maximum increase after six years of 7.3 ppbv. This is due to heterogeneous reactions on the aerosols, leading to H_2O production from methane: Because the stratospheric H_2O in the model is calculated assuming constant total hydrogen, a decrease in methane will increase the amount of H_2O .

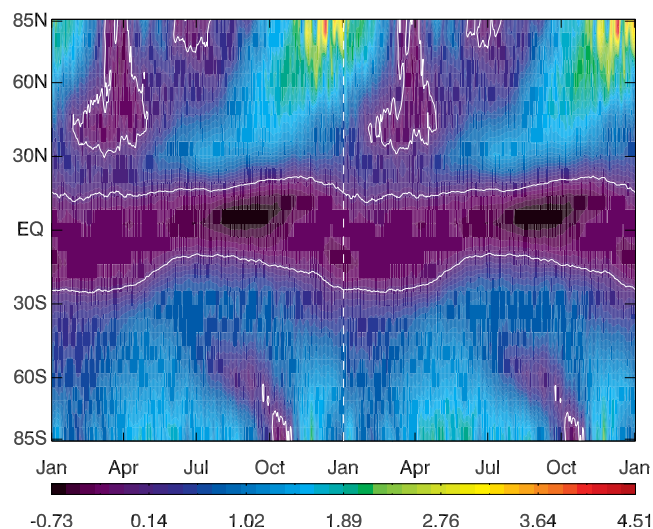


Fig. 15. Changes in the daily zonal mean total ozone column [Dobson Units] due to the mixed fleet aerosol emissions (AERO minus MIX), for the last two years of simulations, ranging from -0.73 DU to 4.51 DU.

5 Discussion and conclusions

We have shown that aerosol emissions in a projected future (2050) “mixed fleet” of subsonic and supersonic aircraft are likely to increase stratospheric ozone due to a lowered loss to NO_x , leading to an increase in the total ozone column. Without aerosol emissions, subsonic aircraft are found to increase tropospheric ozone and lower stratospheric ozone in agreement with previous studies, although with a larger impact (7 to 18 ppbv ozone increase in the monthly zonal mean) compared to previous studies of present impact (3 to 8 ppbv; Gauss et al., 2006a). Similarly, supersonic aircraft are estimated to give ozone depletion in the stratosphere (31 to 49 ppbv as monthly zonal mean). A mixed fleet will therefore reduce the total ozone column compared to a subsonic fleet. Reduced stratospheric ozone in the Southern Hemisphere is a result of cross hemispheric transport of ozone depleted air. At northern high latitudes, aircraft emissions cause an increase in the total ozone column in spring and early summer – when transport processes and the amount of sunlight increase the ozone production – and a decrease in autumn.

The maximum contribution of H_2O emissions from a mixed fleet to stratospheric ozone depletion ranges from 6.7 to 11.4 ppbv in the monthly zonal mean. The largest effect is found to occur through heterogeneous chemistry in October and November.

When aircraft aerosol emissions are introduced, NO_x levels are reduced due to heterogeneous chemistry, while ClO levels increase. This results in enhanced ozone loss in the 11–20 km region. To some extent aircraft aerosols will also

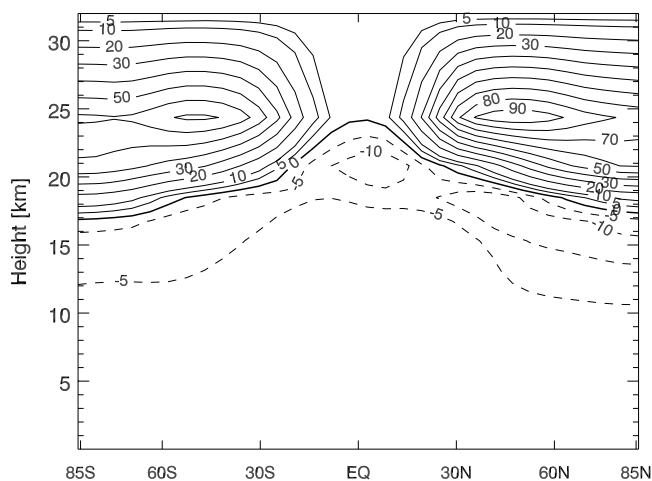


Fig. 16. Annual zonal mean effect of aircraft aerosol emissions on O_3 (AERO minus MIX), given in ppbv.

reduce tropospheric NO_x and therefore ozone production, contributing to a reduction of the total ozone column. The heterogeneous chemistry and the lowered production contributes to a reduction in the total ozone column, whereas the stratospheric NO_x reduction contribute to an ozone column increase.

An increase in the surface area density of aerosols converts more NO_x to HNO_3 and the reduction of NO_x activates more chlorine, and increase ozone loss due to PSCs. Background aerosols are kept at the low 1999 level. Increases in the background aerosols increase, e.g. due to volcanic eruptions, will affect NO_x and the ozone loss. Likewise, adopting the 2000 meteorology as a reference for this study to ensure stable steady-state results, could have an impact on the calculated perturbations. The year 2000 has been reported to exhibit a cold bias near the tropopause (Randel et al., 2004). In a too cold UTLS region the heterogeneous conversions would be overestimated which would lead to a smaller O_3 -loss due to NO_x and larger loss due to halogens. In respect to this, the 2000 UTLS could be more similar to year 2050 conditions with increased CO_2 levels. Assessing the impact of meteorology on the chemical perturbations in the UTLS region is being done in another study.

Although T21 reproduces the main features of measurements, it is a rather coarse horizontal resolution. We find it sufficient for this study, combined with the applied aircraft plume model. Increasing the resolution to T42 will be done in the future, as well as increasing the vertical resolution to cover the whole stratosphere.

It should be noted that we have assumed no sedimentation of aircraft aerosols. If included, the effect of the calculated aerosol surface area density would probably be somewhat reduced, with the result that the ozone reduction due to lowered NO_x and increased ClO would be smaller, possibly

resulting in a decrease in the total ozone column. Nevertheless, aircraft aerosol emissions seem to be an important part of the stratospheric aircraft emissions.

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